SHG without phase-matching

Non-depleted pump approximation: treat A_1 as constant

$$\frac{\partial A_2}{\partial z} = i\frac{\omega_2^2 d}{k_2 c^2} A_1^2 e^{i\Delta k z}$$

Integrate:

$$A_2(L) = i\frac{\omega_2^2 d}{k_2 c^2} A_1^2 \int_0^L e^{i\Delta k z}\, dz$$

$$A_2(L) = i\frac{\omega_2^2 d}{k_2 c^2} A_1^2 L \frac{\left(e^{i\Delta k L} - 1\right)}{i\Delta k L}$$

Convert to intensity $\quad I_2 = 2\varepsilon_0 n_2 c \left|A_2\right|^2$

$$\rightarrow \frac{1}{2\varepsilon_0 n_2 c} I_2(z) = \left(\frac{1}{2\varepsilon_0 n_1 c}\right)^2 I_1^2 \left(\frac{\omega_2 d}{n_2 c}\right)^2 L^2 \left(\frac{\sin(\Delta k L/2)}{\Delta k L/2}\right)^2$$

$$\rightarrow I_2(L) = \frac{\omega_2^2 d^2}{2\varepsilon_0 n_1^2 n_2 c^3} I_1^2 L^2 \operatorname{sinc}^2(\Delta k L/2)$$

As a function of L and fixed |Δk|>0:

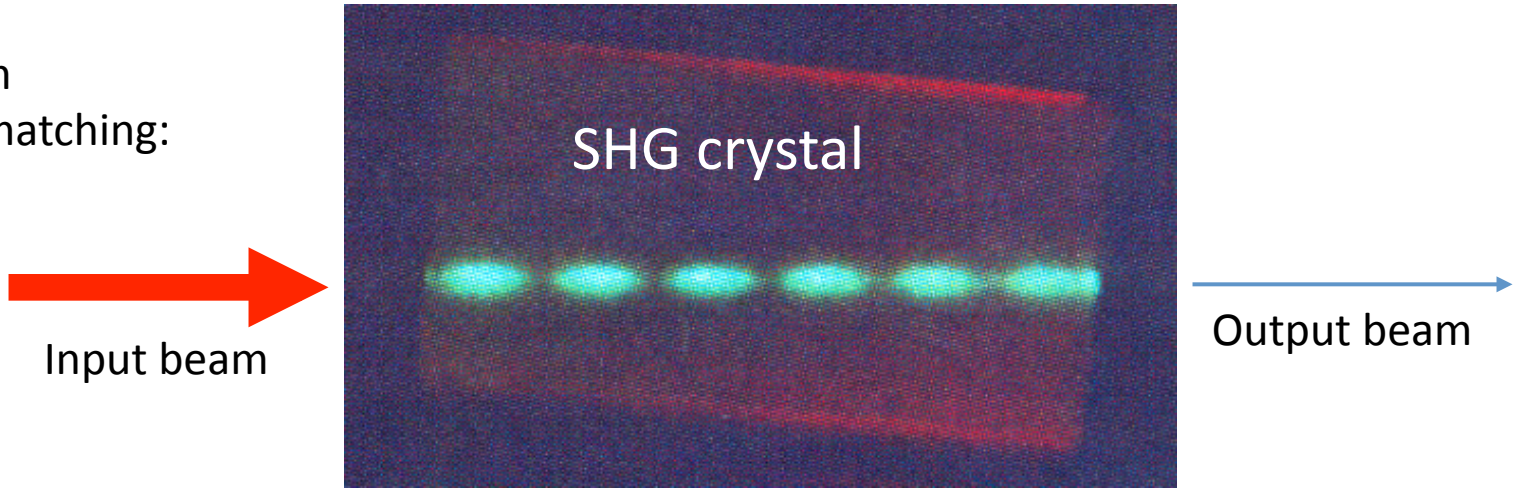
$$I_2(L) = \frac{\omega_2^2 d^2}{2\varepsilon_0 n_1^2 n_2 c^3} I_1^2 \frac{4}{\Delta k^2} \sin^2(\Delta k L/2)$$

Yield oscillates:

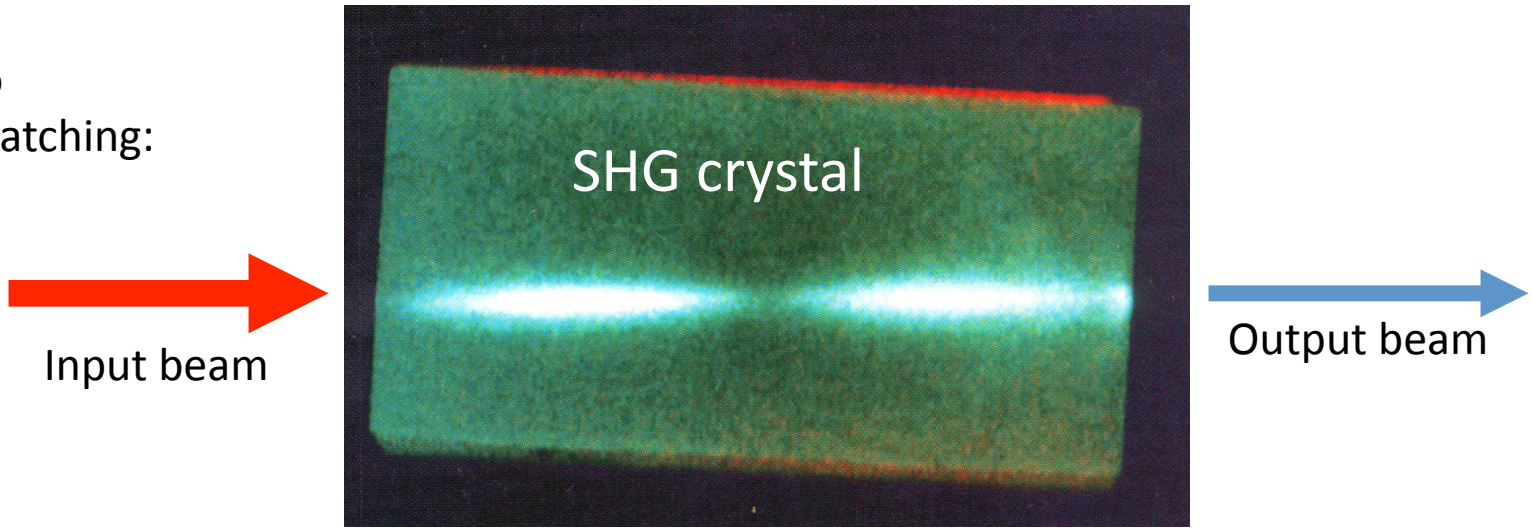
- Period = "coherence length" $\quad L_{coh} = 2\pi/\Delta k$
- Amplitude proportional to $\quad \max(I_2) \propto 1/\Delta k^2$

Light created in real crystals

Far from
phase-matching:



Closer to
phase-matching:



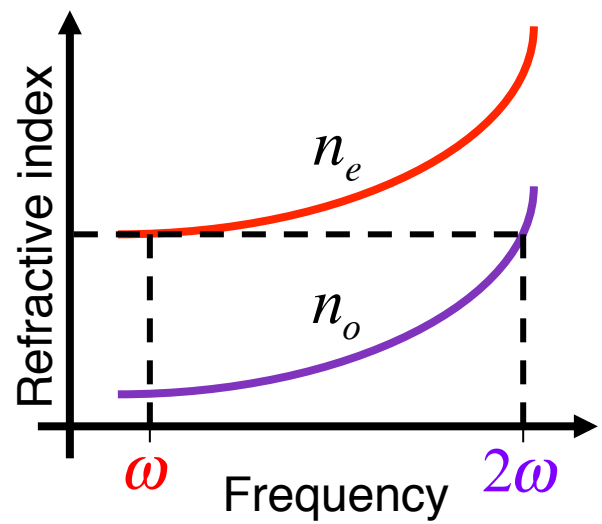
Note that SH beam is brighter as phase-matching is achieved.

Phase-matching Second-Harmonic Generation using birefringence

Birefringent materials have different refractive indices for different polarizations. "Ordinary" and "Extraordinary" refractive indices can be different by up to 0.1 for SHG crystals.

We can now satisfy the phase-matching condition.

Put the highest frequency on the lowest index: for negative uniaxial use the extraordinary polarization for ω and the ordinary for 2ω:



$$n_e(\omega,\theta) = n_o(2\omega)$$

n_e depends on propagation angle, so we can tune for a given ω.

Some crystals have $n_e < n_o$, so the opposite polarizations work.

Real crystal dispersion data

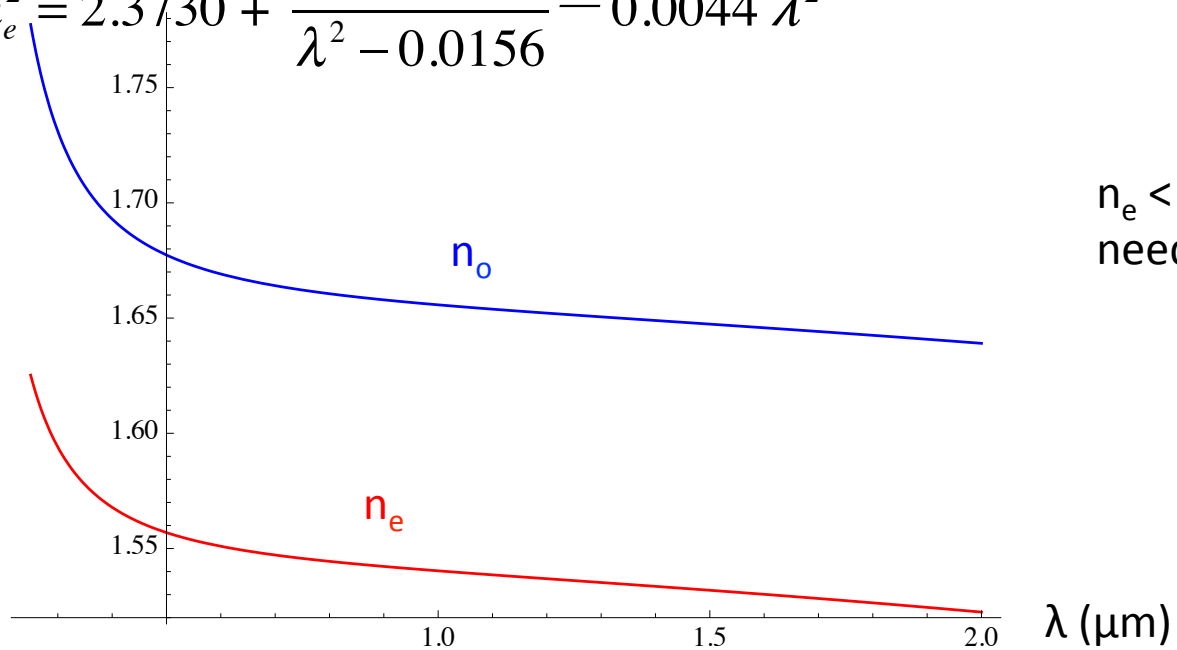
- Best resource: refractiveindex.info
- Others: crystal manufacturers, Handbook of Optics

Example: β-BBO = barium borate, BaB_2O_4

$$n_o^2 = 2.7405 + \frac{0.0184}{\lambda^2 - 0.0179} - 0.0155\,\lambda^2$$

$$n_e^2 = 2.3730 + \frac{0.0128}{\lambda^2 - 0.0156} - 0.0044\,\lambda^2$$

λ is in micrometers!



$n_e < n_o$ everywhere, so we need to angle tune

Types of phase matching

- Type 1:
  - 2ω on low index (n_e)
  - ω on high (n_o)
  - Opposite polarizations ($\chi^{(2)}$ tensor allows this)

$$\Delta k = 2\frac{\omega_1}{c}n_o(\omega_1) - \frac{\omega_2}{c}n_e(\omega_2,\theta)$$
$$= 2\frac{\omega_1}{c}\left(n_o(\omega_1) - n_e(\omega_2,\theta)\right)$$

- Type 2:
  - 2ω on low index (n_e)
  - Project E_1 equally on both axes (n_o and n_e)

$$\Delta k = \frac{\omega_1}{c}n_o(\omega_1) + \frac{\omega_1}{c}n_e(\omega_1,\theta) - \frac{\omega_2}{c}n_e(\omega_2,\theta)$$

- Type 3:
  - "non-critical" or "90°" phase matching
  - Temperature-tuned
  - Only for particular crystals and wavelengths

$$\Delta k = 2\frac{\omega_1}{c}\left(n_o(\omega_1,T) - n_e(\omega_2,90,T)\right)$$

Centro-symmetric media

- For second-order response, the potential must have asymmetry.
- When the binding potential for the electrons is centrally symmetric, the response can still be nonlinear, but the order must be odd (3rd, 5th, etc).
- Consider a central restoring force:

$$\mathbf{F}(\mathbf{r}) = -m\omega_0^2\mathbf{r} + mb(\mathbf{r}\cdot\mathbf{r})\mathbf{r}$$

$$F_i(\mathbf{r}) = -m\omega_0^2 r_i + mbr_j r_j r_i$$

  - force is always directed along $\hat{\mathbf{r}}$ direction
  - At large r, force is less binding.
- As with the non-centrosymmetric potential, perform perturbation expansion.
  - $x^{(2)}$ does not contribute, so $\chi^{(2)}=0$

Solution of 3rd order

- Each term for 1st order solution can be a different frequency

$$\ddot{x}^{(3)} + 2\gamma\dot{x}^{(3)} + \omega_0^2 x^{(3)} = b\left(x^{(1)}\right)^3$$

$$\left(\ddot{x}^{(3)}\left(\omega_q\right) + 2\gamma\dot{x}^{(3)}\left(\omega_q\right) + \omega_0^2 x^{(3)}\left(\omega_q\right)\right)e^{-i\omega_q t} = b\sum_{mnp} x^{(1)}\left(\omega_m\right)x^{(1)}\left(\omega_n\right)x^{(1)}\left(\omega_p\right)e^{-i\left(\omega_m+\omega_n+\omega_p\right)t}$$

  - Note the m, n, p can all be + or – : for example, $\omega_{-2} = -\omega_2$
  - Enforce energy conservation, so

$$\omega_q = \omega_m + \omega_n + \omega_p \rightarrow (mnp) \quad \text{in summation}$$

- Solution is

$$\mathbf{r}^{(3)}\left(\omega_q\right) = -\sum_{(mnp)} \frac{be^3}{m^3} \frac{\left(\mathbf{E}\left(\omega_m\right)\cdot\mathbf{E}\left(\omega_n\right)\right)\mathbf{E}\left(\omega_p\right)}{D\left(\omega_q\right)D\left(\omega_m\right)D\left(\omega_n\right)D\left(\omega_p\right)}$$

$$\mathbf{P}^{(3)}\left(\omega_q\right) = -N\,e\,\mathbf{r}^{(3)}\left(\omega_q\right) = +\sum_{(mnp)} N\frac{be^4}{m^3} \frac{\left(\mathbf{E}\left(\omega_m\right)\cdot\mathbf{E}\left(\omega_n\right)\right)\mathbf{E}\left(\omega_p\right)}{D\left(\omega_q\right)D\left(\omega_m\right)D\left(\omega_n\right)D\left(\omega_p\right)}$$

Calculation of $\chi^{(3)}$

- 3rd order NL polarization is

$$\mathbf{P}^{(3)}\left(\omega_q\right) = \sum_{(mnp)} N \frac{be^4}{m^3} \frac{\left(\mathbf{E}\left(\omega_m\right)\cdot\mathbf{E}\left(\omega_n\right)\right)\mathbf{E}\left(\omega_p\right)}{D\left(\omega_q\right)D\left(\omega_m\right)D\left(\omega_n\right)D\left(\omega_p\right)}$$

- Defined in terms of the susceptibility

$$P_i^{(3)}\left(\omega_q\right) \equiv \varepsilon_0 \sum_{jkl} \sum_{(mnp)} \chi_{ijkl}^{(3)}\left(\omega_q,\omega_m,\omega_n,\omega_p\right) E_j\left(\omega_m\right) E_k\left(\omega_n\right) E_l\left(\omega_p\right)$$

- $\chi^{(3)}$ is a *tensor*:
  - *i j k l* are *coordinate* indices (1, 2, 3 or *x, y, z*) that correspond to the directions of the *field* polarizations: *i* is output, *j, k, l* are distinct inputs
  - q, m, n, p are frequency indices of the distinct fields
  - All indices can potentially be the same
  - (mnp) in summation means $\omega_q = \omega_m + \omega_n + \omega_p$

$\chi^{(3)}$ tensor

- Convert vector P to summation: e.g.

$$\mathbf{E}(\omega_m)\cdot\mathbf{E}(\omega_n)=\sum_j E_j(\omega_m)E_j(\omega_n)=\sum_{jk} E_j(\omega_m)E_k(\omega_n)\delta_{jk}$$

- 3rd order NL susceptibility is

$$P_i^{(3)}(\omega_q)=\sum_{jkl}\sum_{(mnp)} N\frac{be^4}{m^3}\frac{E_j(\omega_m)E_k(\omega_n)E_l(\omega_p)\delta_{jk}\delta_{il}}{D(\omega_q)D(\omega_m)D(\omega_n)D(\omega_p)}$$

$$P_i^{(3)}(\omega_q)\equiv\varepsilon_0\sum_{jkl}\sum_{(mnp)}\chi_{ijkl}^{(3)}(\omega_q,\omega_m,\omega_n,\omega_p)E_j(\omega_m)E_k(\omega_n)E_l(\omega_p)$$

$$\chi_{ijkl}^{(3)}(\omega_q,\omega_m,\omega_n,\omega_p)=\frac{Nbe^4}{\varepsilon_0 m^3}\frac{\delta_{jk}\delta_{il}}{D(\omega_q)D(\omega_m)D(\omega_n)D(\omega_p)}$$

- Account for "intrinsic permutation symmetry"
  - Fields $E_j(\omega_m)E_k(\omega_n)E_l(\omega_p)$ can be in any order

$$\chi_{ijkl}^{(3)}(\omega_q,\omega_m,\omega_n,\omega_p)=\frac{Nbe^4}{3\varepsilon_0 m^3}\frac{\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}}{D(\omega_q)D(\omega_m)D(\omega_n)D(\omega_p)}$$

  - 3 terms aren't there b/c of dot product of fields

NL polarization: 2nd order

$$P_i^{(2)}(\omega_n+\omega_m)=\varepsilon_0\sum_{jk}\sum_{(nm)}\chi_{ijk}^{(2)}(\omega_n+\omega_m,\omega_n,\omega_m)E_j(\omega_n)E_k(\omega_m)$$

In this situation, each χ(2) is a 3x3 matrix, and the vector form would be written

$$P_i^{(2)}(\omega_n+\omega_m)=\varepsilon_0\sum_{(nm)}\mathbf{E}(\omega_n)\bullet\overleftrightarrow{\chi}_i^{(2)}(\omega_n+\omega_m;\omega_n,\omega_m)\bullet\mathbf{E}(\omega_m)$$

In matrix form, this would be

$$P_1^{(2)}(\omega_3=\omega_1+\omega_2)=\varepsilon_0\begin{pmatrix} E_1(\omega_1) & E_2(\omega_1) & E_3(\omega_1)\end{pmatrix}\begin{pmatrix}\chi_{111} & \chi_{112} & \chi_{113}\\ \chi_{121} & \chi_{122} & \chi_{123}\\ \chi_{131} & \chi_{132} & \chi_{133}\end{pmatrix}\begin{pmatrix}E_1(\omega_2)\\ E_2(\omega_2)\\ E_3(\omega_2)\end{pmatrix}$$

$$+\varepsilon_0\begin{pmatrix} E_1(\omega_2) & E_2(\omega_2) & E_3(\omega_2)\end{pmatrix}\begin{pmatrix}\chi_{111} & \chi_{112} & \chi_{113}\\ \chi_{121} & \chi_{122} & \chi_{123}\\ \chi_{131} & \chi_{132} & \chi_{133}\end{pmatrix}\begin{pmatrix}E_1(\omega_1)\\ E_2(\omega_1)\\ E_3(\omega_1)\end{pmatrix}$$

We need to use as many symmetries as possible to reduce the complexity.

can reduce from ~ 324 terms to 10 or fewer

Summary of intrinsic symmetries for NL tensor

- Real fields and polarization

$$E_j(-\omega_n) = E_j^*(\omega_n) \quad P_i(-\omega_m - \omega_n) = P_i^*(\omega_m + \omega_n)$$

$$P_i^{(2)}(\omega_n + \omega_m)^* = \varepsilon_0 \sum_{jk} \sum_{(nm)} \chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m)^* E_j(\omega_n)^* E_k(\omega_m)^*$$

$$P_i^{(2)}(-\omega_n - \omega_m) = \varepsilon_0 \sum_{jk} \sum_{(nm)} \chi_{ijk}^{(2)}(-\omega_n - \omega_m, -\omega_n, -\omega_m) E_j(-\omega_n) E_k(-\omega_m)$$

- Intrinsic permutation symmetry:
  - j, k and m, n are dummy indices

$$\chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m) E_j(\omega_n) E_k(\omega_m) = \chi_{ikj}^{(2)}(\omega_n + \omega_m, \omega_m, \omega_n) E_k(\omega_m) E_j(\omega_n)$$

  - Swap j k and n m at same time. Not i and q in this case

Other common symmetries

- Lossless media (not always true)
  - Real components of $\chi^{(2)}$. Usually true if incident frequencies and their combinations are away from resonances.
  - full permutation symmetry: free interchange of all ω components, as long as i, j, k are swapped at same time.

- Kleinmans's symmetry (if dispersion of $\chi^{(2)}$ can be neglected)
  - Dispersion is intrinsically linked to time response. So if we neglect dispersion, it is the same as assuming NL response is instantaneous.
  - In this case, we can permute I j k without also permuting ω's
  - This makes χ tensors symmetric
- **Note:**
  - From practical perspective of using NL crystals, many desirable crystals are designed to be transparent, non-dispersive, etc.
  - But for characterizing materials, measuring NL response leads to important information about structure and internal energy levels

Contracted notation: non-dispersive, non-absorbing medium

Non-dispersive: can permute any spatial index (Kleinmann symm)
- like colors have the same value, except black terms are unique

$$\begin{pmatrix} d_{xxx} & d_{xxy} & d_{xxz} \\ & d_{xyy} & d_{xyz} \\ & & d_{xzz} \end{pmatrix}$$

$$\begin{pmatrix} d_{xxy} & d_{xyy} & d_{xyz} \\ & d_{yyy} & d_{yyz} \\ & & d_{yzz} \end{pmatrix}$$

$$\begin{pmatrix} d_{xxz} & d_{xyz} & d_{xzz} \\ & d_{yyz} & d_{yzz} \\ & & d_{zzz} \end{pmatrix}$$

$$\begin{pmatrix} d_{xxx} & d_{xyy} & d_{xzz} & d_{xyz} & d_{xxz} & d_{xxy} \\ d_{xxy} & d_{yyy} & d_{yzz} & d_{yyz} & d_{xyz} & d_{xyy} \\ d_{xxz} & d_{yyz} & d_{zzz} & d_{yzz} & d_{xzz} & d_{xyz} \end{pmatrix}$$

$$\begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{14} & d_{12} \\ d_{15} & d_{24} & d_{zzz} & d_{23} & d_{13} & d_{14} \end{pmatrix}$$

Spatial symmetries

- Crystal structure leads to spatial symmetries: 32 different crystal point groups.
- Example: 4-fold symmetry around z-axis means $\chi_{zxx}=\chi_{zyy}$
- Spatial symmetry affects
  - linear optical properties (birefringence, optical activity)
  - high-order χ tensors
  - Even if $\chi^{(1)}$ is isotropic, $\chi^{(3)}$ may not be. Ex: XPW generation
- Inversion symmetry and $\chi^{(2)}$

$$P(t)=\varepsilon_0\chi^{(2)}E^2(t)$$

  - If medium is centrosymmetric (possesses inversion symmetry), then P(t) must have same sign as E(t)

$$-P(t)=\varepsilon_0\chi^{(2)}\left(-E(t)\right)^2$$

  - This means that $\chi^{(2)}$ must vanish.

Symmetry in d-matrices

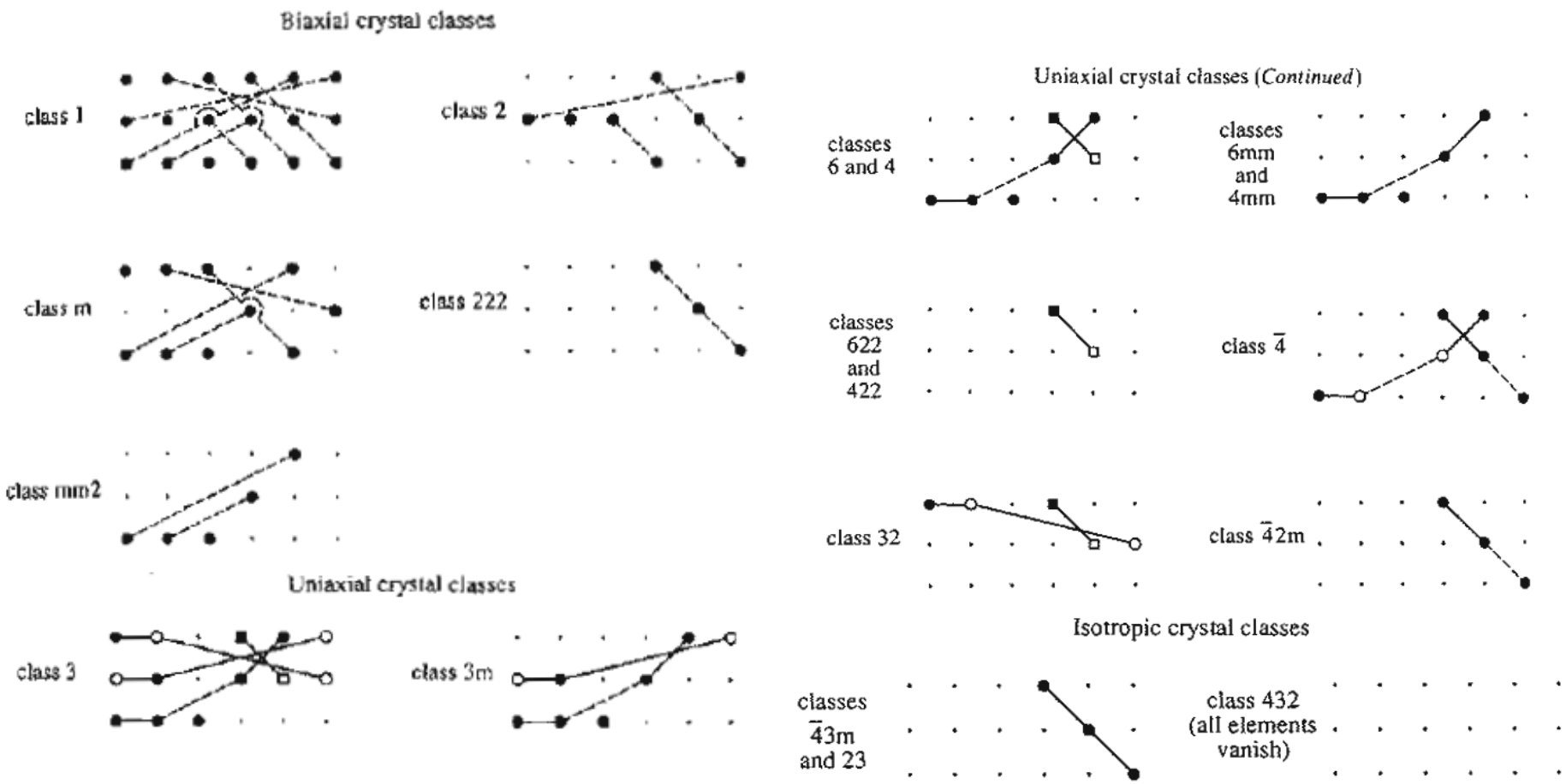


FIGURE 1.5.3 Form of the d_{ij} matrix for the 21 crystal classes that lack inversion symmetry. Small dot: zero coefficient; large dot: nonzero coefficient; square: coefficient that is zero when Kleinman's symmetry condition is valid; connected symbols: numerically equal coefficients, but the open-symbol coefficient is opposite in sign to the closed symbol to which it is joined. Dashed connections are valid only under Kleinman's symmetry conditions. (After Zernike and Midwinter, 1973.)

Common NL crystals

- KDP, KD*P:
  - uniaxial, can grow large crystals, low dispersion.
  - Doubling, OPA Pockels cells,…
- BBO:
  - uniaxial, high NL coeff good UV transmisson,
  - Doubling, OPA, Pockels cells…
- KTP:
  - biaxial, high NL coeff for typeII doubling
- LBO:
  - biaxial, high damage threshold
- $LiNbO_3$:
  - pockels cells, PPLN
- Newer: BiBO, ZGP
- Suppliers: Casix, Castech, EKSMA, Quantum Tech, Cleveland Crystals…

Evaluating crystals for applications

- Transparency for spectral region
- Orientations that allow for NL response (tensors), strength of NL coefficient
- Phase-matching geometries allowed
- Damage threshold (max intensity)
- Thermal issues (change in index with temperature)
- CW: birefringent walk-off (sets limit for crystal length)
- Angular acceptance: limits divergence of input, crystal length
- Short pulses: dispersion/phase matching bandwidth, connected to group velocity walkoff
- Dimensions of crystal, cost